Karlsruhe Institute of Technology

FIRST Nuclides

FIRST-Nuclides: Outcome, Open Questions and Steps Forward

IGD-TP Exchange Forum n°5, October 28-30th, 2014, Kalmar, Sweden

Bernhard Kienzler, KIT-INE, Germany

Institut für Nukleare Entsorgung (INE)

Subatech

Hungarian Academy of Sciences, Centre for Energy Research

Acknowledgement: The research leading to these results has received funding from the European Atomic Energy Community's Seventh Framework Programme (FP7/2007-2011) under grant agreement no. 295722, the FIRST-Nuclides project.

Associated Groups

13 Groups participating at their own costs with specific RTD contributions or particular information exchange functions, or mobility measures.

End User Group

Review the scientific-technical progress and contributions to the proceedings and assess the project status and provide recommendations for the work program

Objectives

- Experimental
  - Quantification of relation FGR to IRF for ^{129}I, ^{79}Se, ^{135}Cs
    - for relevant burn-up / lin. power rate ranges,
    - for full set of sample sizes (pellet, ..., powder),
    - for same groundwater different atmospheric conditions
  - If possible: Quantification (speciation) of elements such as ^{14}C, Se
- Modelling
  - Up-scaling
- Training, Education, Dissemination
- Further expected outcome of FIRST-Nuclides
  - Definition of instant – rapid – fast release and delineation to long-term RN release processes.
  - Up-scaling from lab sample to rod/assembly.
  - Effect of geochemical conditions.

Materials

Selection, characterization and preparation of materials and set-up of tools

- Characterization of selected SNF materials with respect to
  - Fuel characteristics, irradiation history,
  - Permission by the fuel owner for publication of key parameters
- Preparation of SNF samples, cladded pellets, pellets, powders, etc.
- Experimental set-ups (autoclaves, irradiation cells, reaction vessels),
  - Sampling devices
  - Analytical equipment in the hot cells

Selected high burn-up UO_2 fuel samples

FIRST Nuclides

Relevant SNF samples ✓

	Units	PWR	BWR	THTR / VVER
Enrichment	%	**3.80 – 4.94 %**	**3.30 - 4.25 %**	2.4 -16.8%
Burn-up	GWd/t_{HM}	**50.4 – 70.2**	**48.3 – 57.5**	
lin. power	W/cm	**186 - 330**	**160**	130 – 228
FGR		**4.9 – 23 %**	**1.2 – 3.1 %**	

WP 2: Gas release and rim and grain boundary diffusion

Direct comparison between FGR and IRF ✓

Radial FP distrbution over the pellet radius by laser ablation mass spectroscopy ✓

WP 2: Non LWR materials

IRF from spent UO2 TRISO coated particles, microstructure evolution ✓

WP leader: Detlef Wegen (JRC-ITU)

Results on speciation of Selenium

FIRST Nuclides

XANES measurements: mixture Se(0) and Se(IV) or pure Se(-II)

Modelling: Homogeneous chemical form, probably as Se(-II) replacing oxygen sites in the UO_2 lattice.

Cooperation PSI & Studsvik

✓

Curti E. et al. (in press). Selenium redox speciation and coordination in high-burnup UO_2 fuel: Consequences for the release of ^{79}Se in a deep underground repository. J. Nucl. Mat., DOI: 10.1016/j.jnucmat.2014.07.003.

Behaviour of ^{14}C

Dissolution based measurements:

➔ **IRF(^{14}C): below 1.5%** ✓

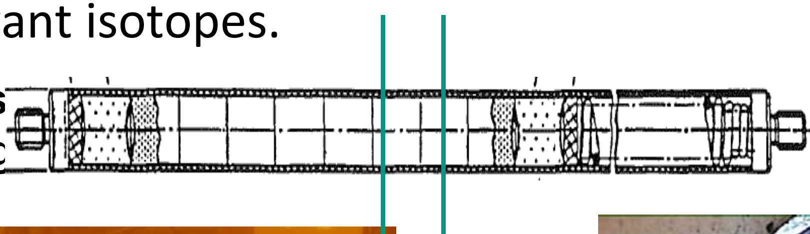
^{14}C in the plenum gas measurement: ✓

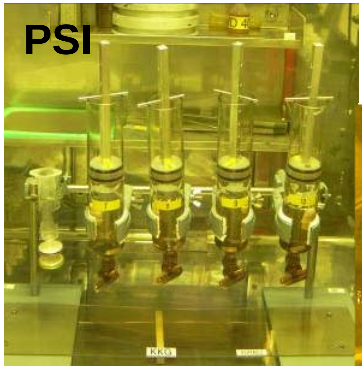
➔ **IRF($^{14}CO_2$ gas) = 0.2 % of ^{14}C inventory**

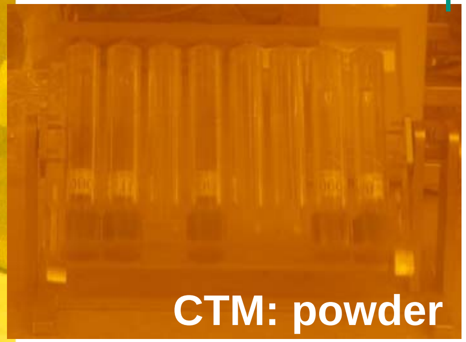
WP 3: Dissolution based release

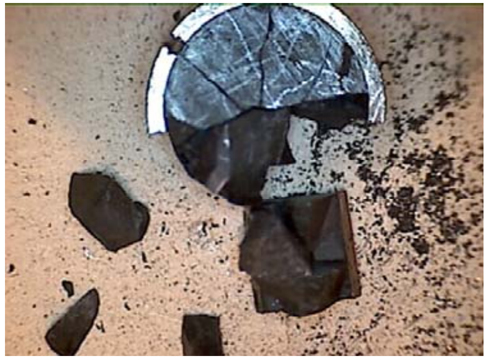
- Dissolution based radionuclide release and to the extent possible the chemical speciation of the relevant isotopes.

powder, fragments
pellets +/- cladding







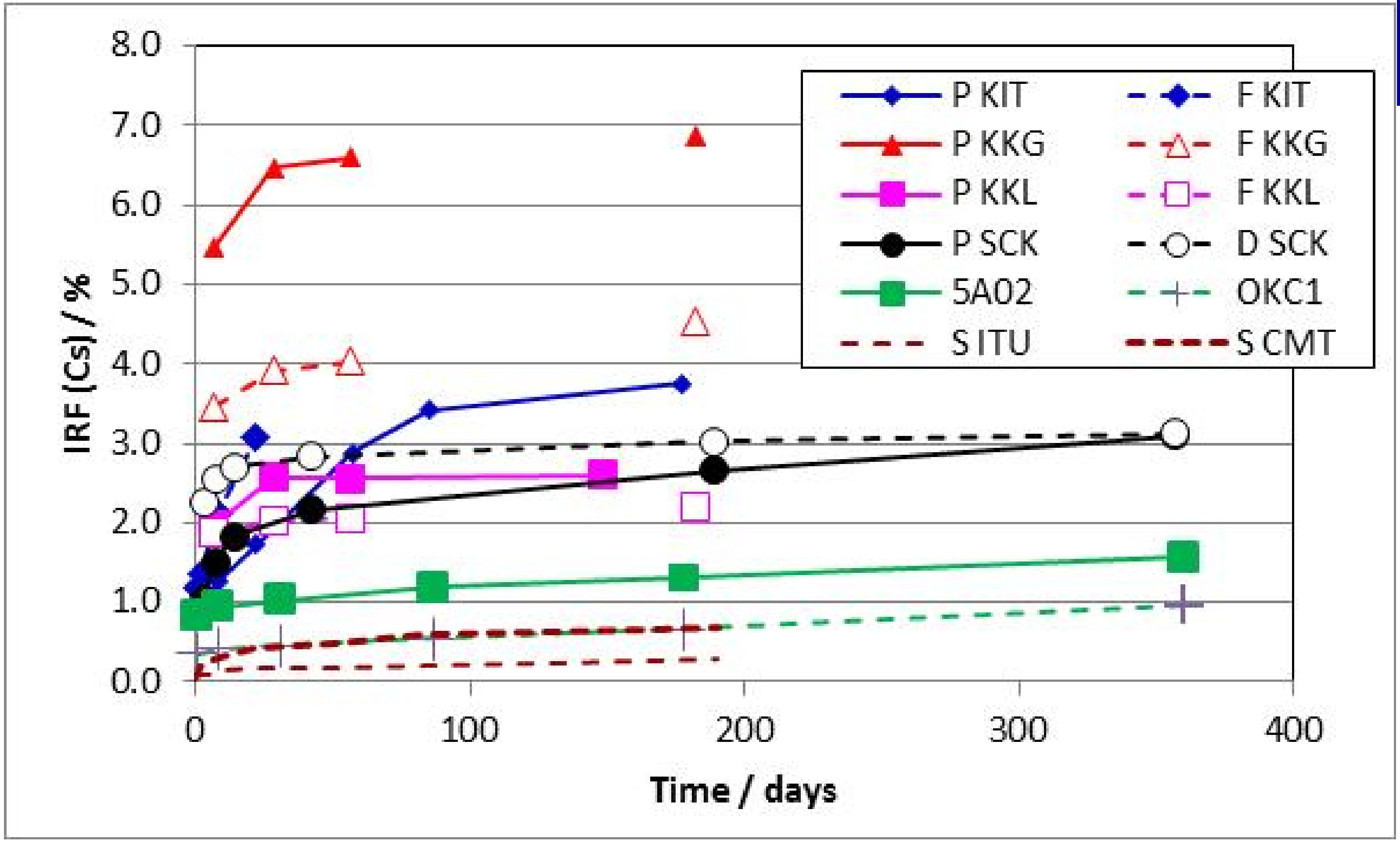


Leachants

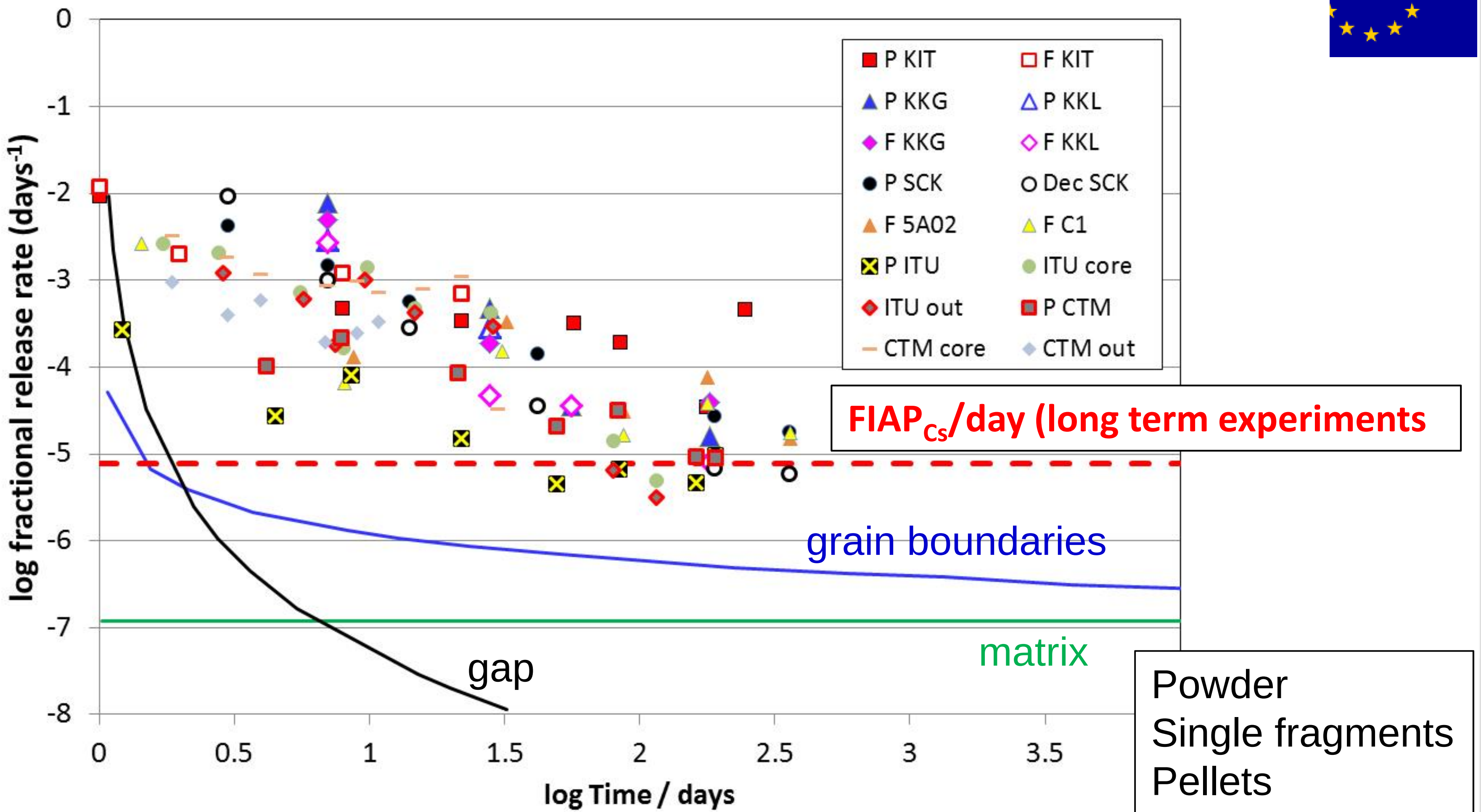
	NaCl	$NaHCO_3^-$	pH	Conditions
PSI	19	1	8.5	anoxic / slightly oxidizing
KIT	19	1	8.9	Ar/H_2 (Eh = -116 mV)
ITU/CTM	19	1	7.4	Oxidizing (air)
SCK·CEN	19	1		anoxic / slightly oxidizing
Studsvik	10	2	8.1-8.2	Oxidizing (air)

WP leader: Karel Lemmens (SCK·CEN)

IRF$_{Cs}$ of different fuel / sample sizes

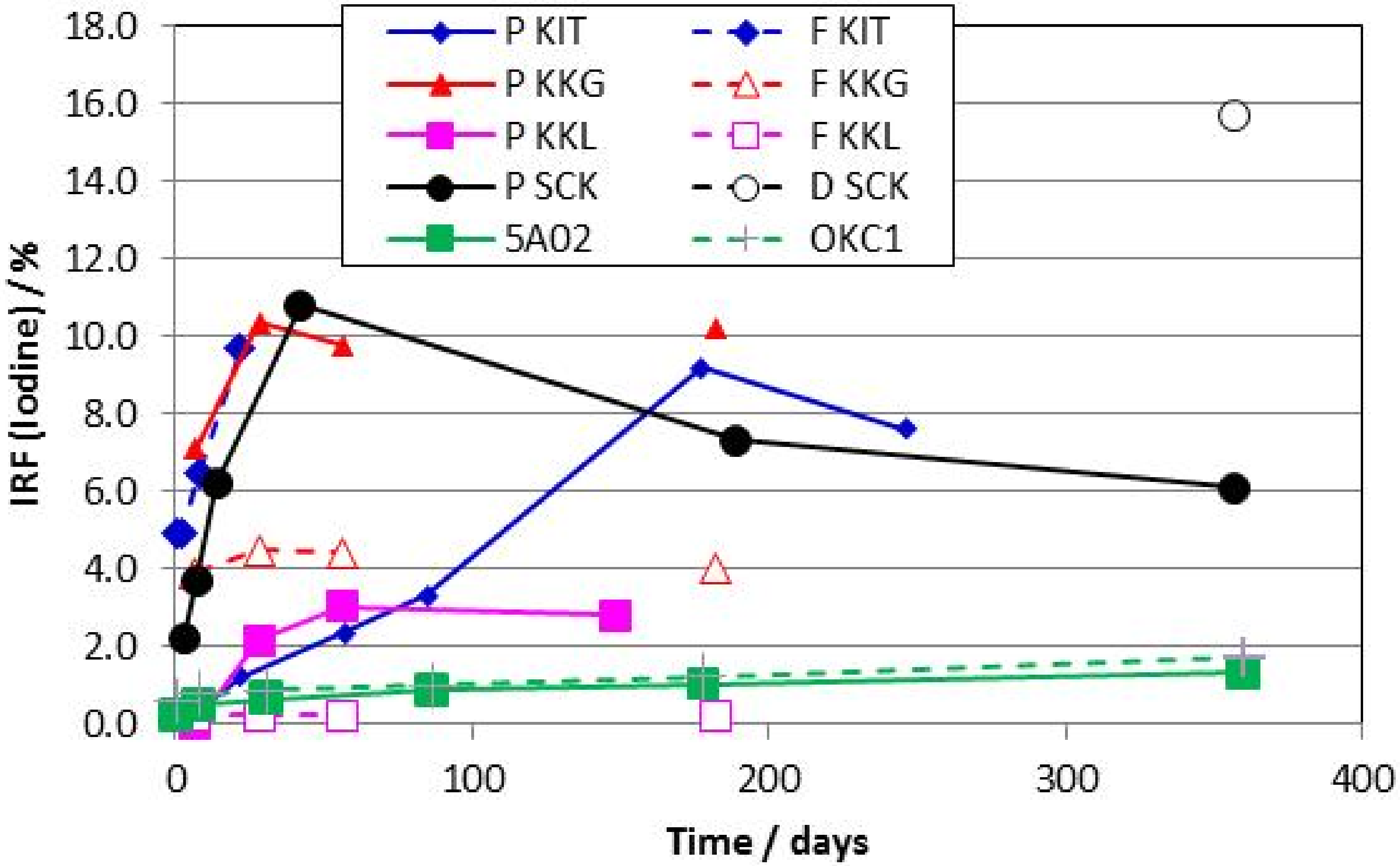


IRF Cs: Delineation from long-term release

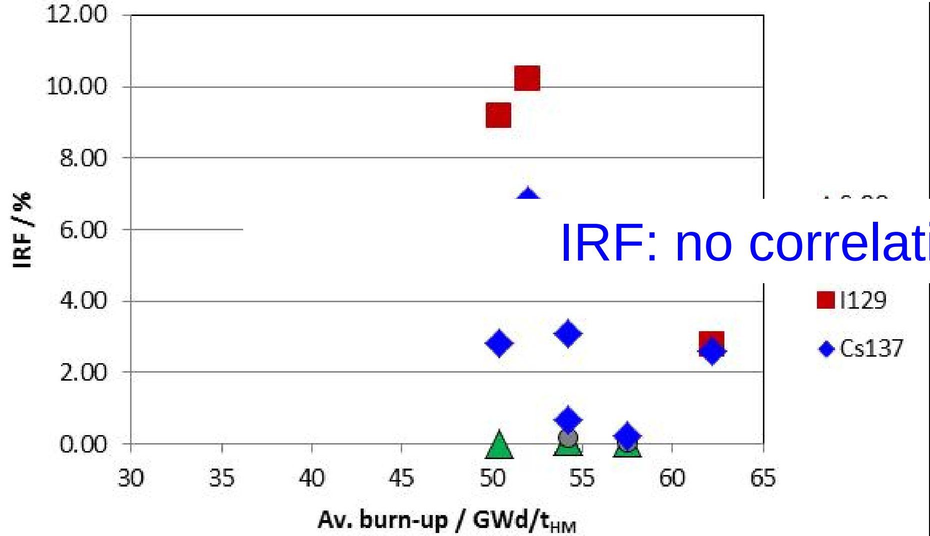


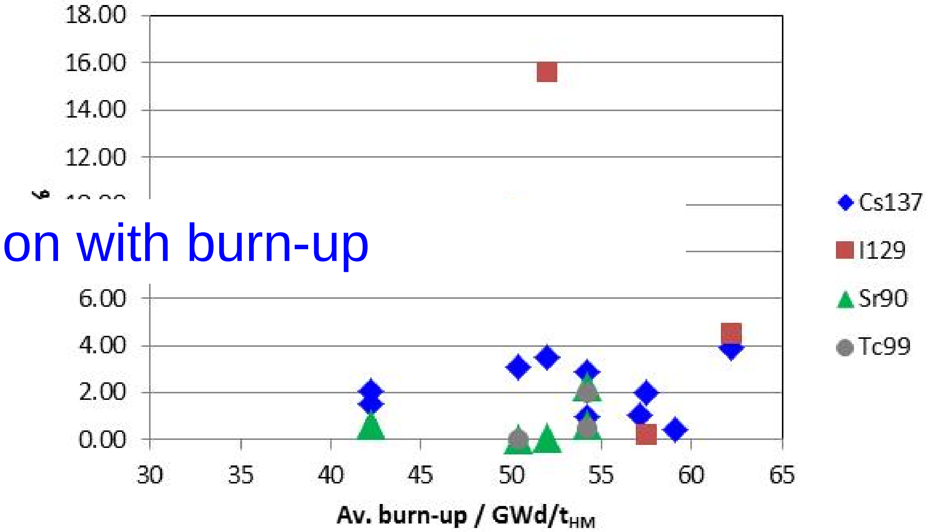
Powder
Single fragments
Pellets
Slices of pellets

IRF$_{iodine}$ of different fuel / sample sizes



IRF as function of burn-up / linear power

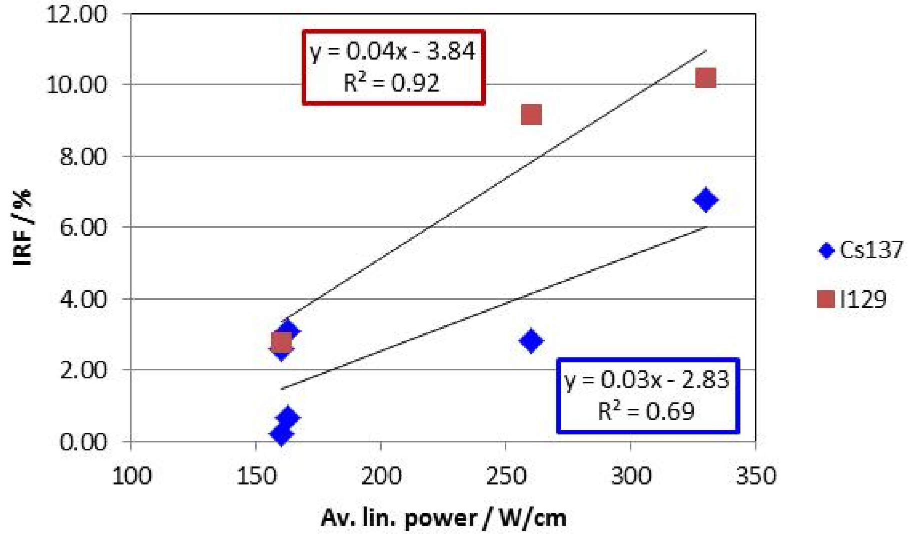


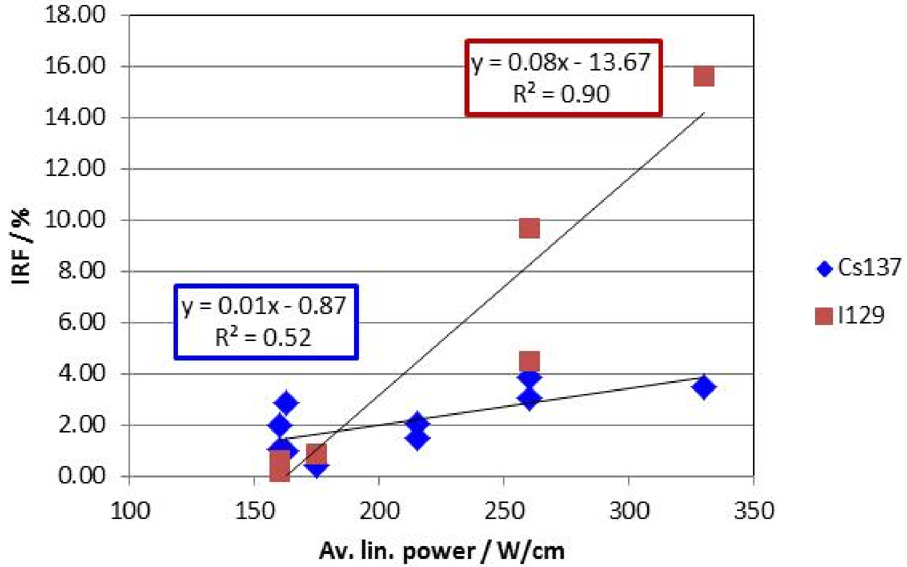


IRF: no correlation with burn-up

Pellets

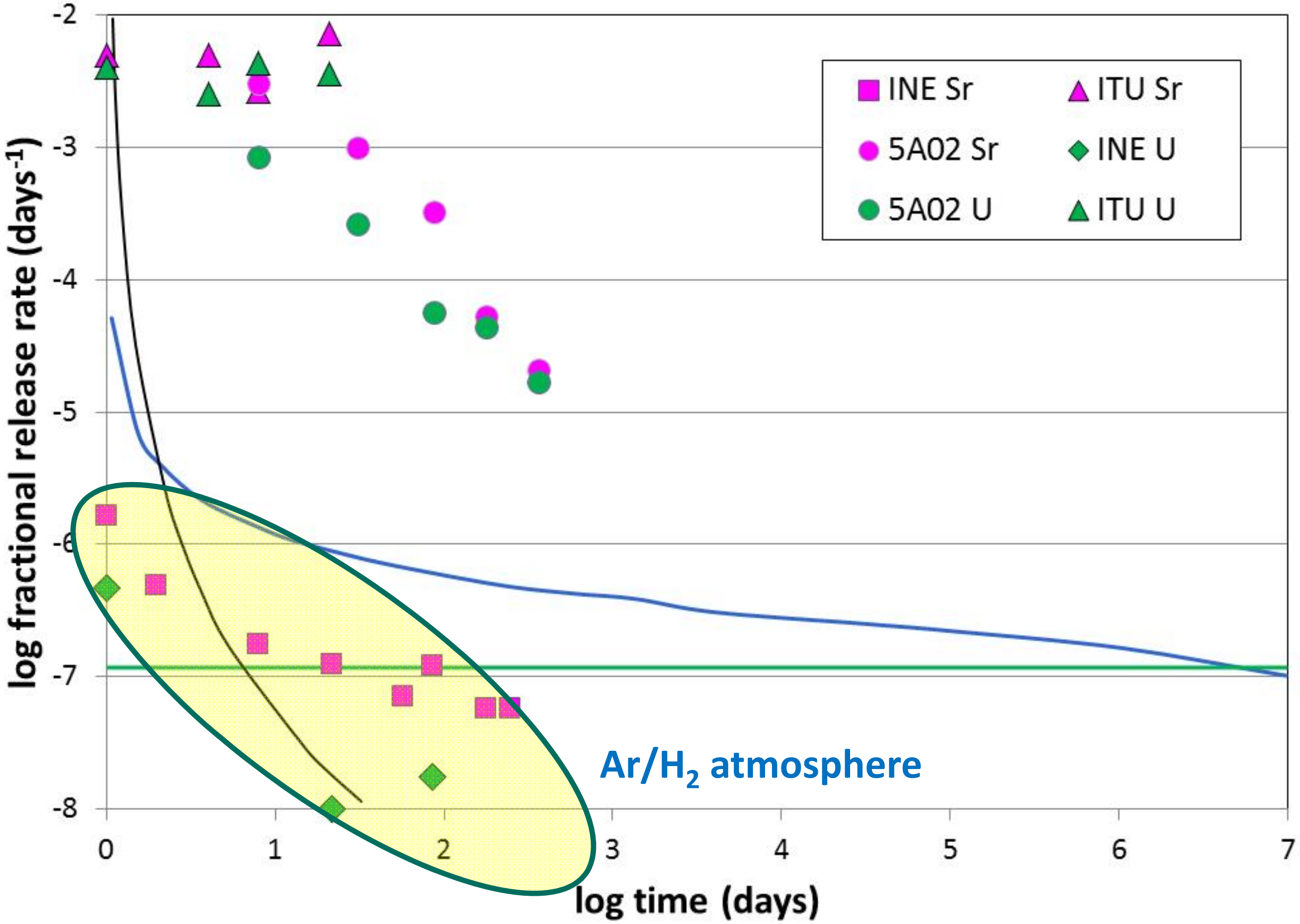
Uncladded, fragments,





IRF: 150-180 days

Effect of reducing conditions on IRF Rates (Sr, U)



Dissemination

1st Annual Workshop Proceedings of the Collaborative Project 'FIRST-Nuclides'
KIT SR 7639 (2013) Download:
http://dx.doi.org/10.5445/KSP/1000032486

2nd Annual Workshop Proceedings of the Collaborative Project 'FIRST-Nuclides'
KIT SR 7676 (2014)

Geodisposal 2014

<u>Training Courses:</u>
Topical sessions during the AWS
Young Scientists Training Course, July 09 – 10, 2013
Young Scientists Mobility Measures

<u>Involvement of non-scientific stakeholders:</u>
Meeting with InSOTEC representatives, July 2013
Presentation of InSOTEC outcome @ 2nd AWS

<u>27th Spent Fuel Workshop:</u>
9 oral talks of FIRST-Nuclides results

Findings

- Release depends on sample characteristics:
  - on the operation parameters, burnup and power rate
  - on the nature of the exposed fuel structures (gap or grain boundaries).
  - Release tends to increase in the order: fragments < clad pellet segments < opened clad pellets, (exposed surface area / gap inventory.
  - Al/Cr doped fuel have lower IRF (larger grain size)
  - IRF higher for PWR fuel as for BWR fuel (linear power).
- Effect of redox conditions for U, Sr, Tc
  - Significant differences for U, Sr and Tc under oxidizing or reducing conditions
- $IRF_{Se/14C}$: few data available presently. Measurements not yet completed.

Evaluation of data obtained in FIRST-Nuclides not yet completed.

End-User Comments

- FIRST-Nuclide is highly relevant for all WMO
- IRF contributes substantially to the peak releases after container breaching and to potential radiological consequences.
- Results include:
  - Data from experimental determination of rapid release fractions for moderate and high burn-up UO_2 fuels.
  - Doped fuels, expected to be used much more in the future.
  - Increased data base for release of Cs and I from high burn-up fuel
  - Comprehensive comparisons of IRF with fission gas release (FGR)
  - Basis to estimate IRF data for a very large number of fuel rods and various reactor operation conditions.
  - Improvement of analytical techniques

Conclusions

- Successful project
- Improved understanding of “IRF“
- Correlations between “reactor data” and IRF
- Publications in preparation
- Huge investments on
  - setting-up the experiments,
  - implementing the required analytical tools and instruments and
  - clearance by the utilities to publish the spent fuel data.
  - Experimental duration could be extended
- Open issues still exist

Steps Forward

- Continuation of the 3 years project
  ➔ maximize the outcome
  ➔ maximal exploitation of investments (hot cell installations, analytics, staff, …)
  - Improved statistics for the IRF of other fission products.
  - In depth investigations of low concentrated but relevant isotopes ^{79}Se, and ^{107}Pd, or activation products ^{36}Cl and ^{14}C.
  - Experiments under repository relevant conditions, i.e. reducing conditions, H2 overpressure
  - Correlations for predicting the IRF from nuclear power plant operational parameters (power rates, ramps, temperatures, FGR).
  - Delineating the instant release from long-term radionuclide release.
  - Additional kind of samples (e.g. MOX, reprocessed uranium fuel, low-FGR fuels, …)
  - Keeping the know-how.

Proposed Procedure

- Keeping the experimental set-up and the materials for a certain interim period in the labs
- Depending on financial support by the WMO
  - Less frequent sampling
  - Storage of samples
- Required support by the WMO/IGD-TP:
  - New project at the next EU Call
  - Materials from utilities (transports, etc.)
  - Receiving operational parameters from the utilities such as power rates, ramps, temperatures, FGR
    - Not only average values!